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Intercomparison of Plutonium-239 Measurements

U.S. Environmental Monitoring and Support Lab, Las Vegas, Nev

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Las Vegas NV 89114

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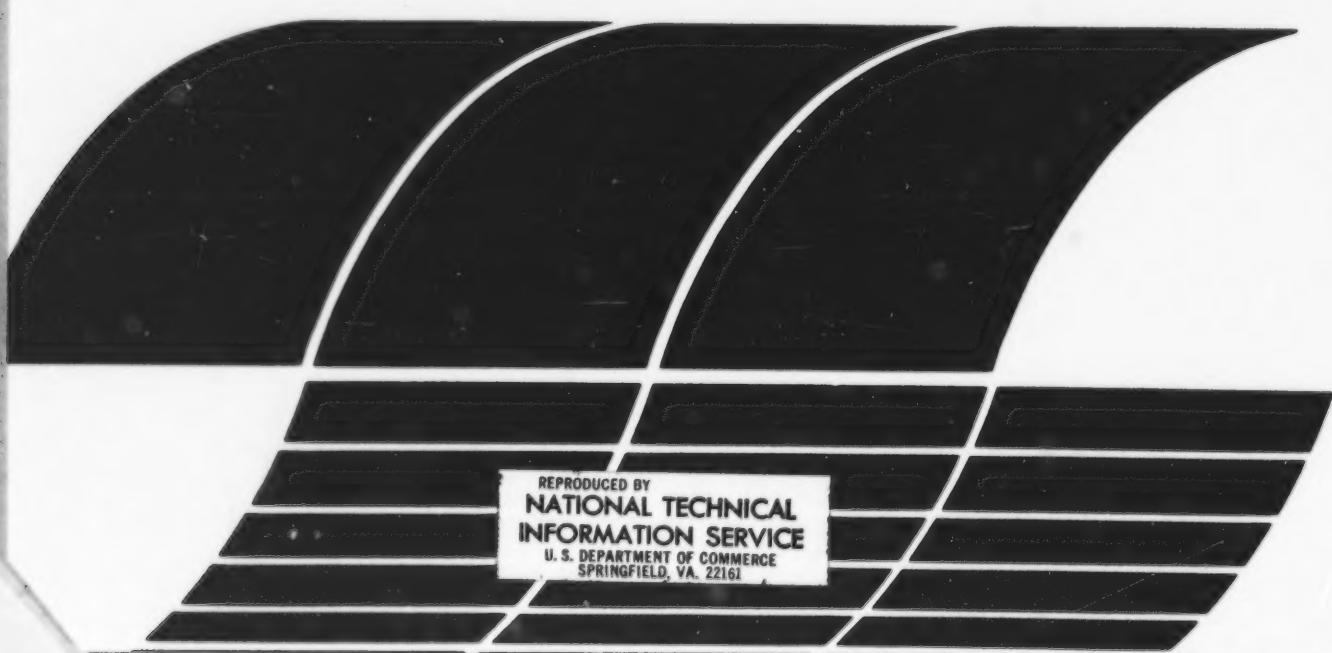
Research and Development

PB 285 438



Intercomparison of Plutonium-239 Measurements

**Interagency
Energy-Environment
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16. ABSTRACT In 1977 the U.S. Environmental Protection Agency distributed calibrated solutions of plutonium-239 to laboratories interested in participating in an intercomparison study of plutonium analysis. Participants were asked to perform a quantitative radioactivity analysis of the solution. The results reported by all the participating laboratories are given here. Eighty-eight percent of the reported activity values were within ± 20 percent of the activity value certified by the Environmental Protection Agency.		
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INTERCOMPARISON OF
PLUTONIUM-239 MEASUREMENTS

by

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OFFICE OF RESEARCH AND DEVELOPMENT
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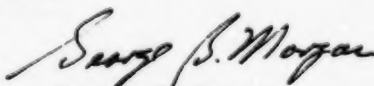
This report has been reviewed by the Environmental Monitoring and Support Laboratory-Las Vegas, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

FOREWORD

Protection of the environment requires effective regulatory actions which are based on sound technical and scientific information. This information must include the quantitative description and linking of pollutant sources, transport mechanisms, interactions, and resulting effects on man and his environment. Because of the complexities involved, assessment of specific pollutants in the environment requires a total systems approach which transcends the media of air, water, and land. The Environmental Monitoring and Support Laboratory-Las Vegas contributes to the formation and enhancement of a sound monitoring data base for exposure assessment through programs designed to:

- develop and optimize systems and strategies for monitoring pollutants and their impact on the environment
- demonstrate new monitoring systems and technologies by applying them to fulfill special monitoring needs of the Agency's operating programs.

This report describes an interlaboratory study measuring the activity of aqueous solutions of plutonium-239. The study was funded under the 17-agency Federal Energy/Environment Research and Development Program. The data from these measurements are useful in evaluating the state of the art of existing measurement techniques which in turn provides valuable input into the establishment of standards and guides, and enforcement activities. The Quality Assurance Branch at the U.S. Environmental Protection Agency's Environmental Monitoring and Support Laboratory in Las Vegas encourages the development and implementation of quality control procedures at all levels of sample collection, analysis, data handling and reporting of environmental radiation measurements and can provide further assistance upon request.



George B. Morgan
Director

Environmental Monitoring and Support Laboratory
Las Vegas

INTRODUCTION

The Quality Assurance Branch at the Environmental Monitoring and Support Laboratory in Las Vegas, Nevada, has maintained an active quality assurance program for the last five years. One major activity of the branch in this program has been repetitive interlaboratory intercomparison studies of radioactivity measurements of environmental media (1, 2, 3, 4, 5). Another major activity of the branch has been the calibration and distribution of calibrated radioactivity samples (6). Over 150 State laboratories, Federal laboratories, commercial environmental monitoring laboratories, national laboratories of other countries, nuclear power facilities, and university laboratories have participated in these programs during this time.

The Quality Assurance Branch in Las Vegas has maintained a continuing intercomparison studies program with the National Bureau of Standards throughout its existence (7). One phase of this program has been the occasional distribution of calibrated radioactivity standards to laboratories participating in the Quality Assurance Branch's programs. A previous distribution of an aqueous polonium-210 solution was done as an intercomparison study (8). This report describes an intercomparison study on measurement of activity of aqueous solutions containing plutonium-239.

These intercomparison studies provide information about the accuracy that monitoring laboratories produce when measuring the activities of aqueous solutions. Also, they are intended to survey the methodology and equipment currently used by these monitoring laboratories.

EXPERIMENTAL

In April, 28 calibrated solutions of plutonium-239 were distributed as unknowns to 28 laboratories which had agreed to participate in an intercomparison study (Appendix). The calibrated solutions were prepared by gravimetric dilutions of a plutonium standard obtained from a commercial vendor. Since the dilutions were made in 100-ml volumetric flasks, aliquots of two dilutions were distributed. One dilution had an activity on December 1, 1973 of 18.63 disintegrations per second per gram of solution, while the other dilution had an activity of 18.46 disintegrations per second per gram of solution. The impurities, expressed as a percentage of the activity of the plutonium-239 on December 1, 1973, were:

Plutonium-238 + Americium-241	(alpha)....0.5%
Plutonium-240	(alpha)....3.62%
Plutonium-241	(beta)....<11%

The activities of the impurities were not included in the activities given above for the plutonium-239 solutions. Consequently, the total alpha-particle emission rate of the calibrated solution would then be 1.04 times the certified activity value.

Besides the calibrated solution, the laboratories were sent one alpha-particle test solution prepared by the National Bureau of Standards with an accompanying report of calibration. A form on which laboratories could report their results and describe the methodology used was also sent to the participating laboratories. This form is similar to that used by the National Bureau of Standards (7) and was used in a previous study (8). The alpha-particle test solution was intended to be used as a calibration standard by the individual laboratories.

By early August, 17 of the laboratories had measured the activity of the plutonium-239 unknown and reported their results. In late August, Calibration Certificates prepared by the Environmental Protection Agency (6) for the calibrated plutonium-239 unknown were sent to all 28 laboratories. The 17 laboratories who returned results are listed in Table 1.

Fifteen laboratories reported that they looked for impurities using spectroscopy. One laboratory also measured the beta activity of the solution. Six laboratories indicated that they observed plutonium-238 in the range of 0.1 nuclear transformations per second per gram ($\text{nts}^{-1}\text{g}^{-1}$) to $0.4 \text{ nts}^{-1}\text{g}^{-1}$, in general agreement with the certified value of 0.5% or $0.09 \text{ nts}^{-1}\text{g}^{-1}$. One laboratory indicated they observed plutonium-242, but they did not quantify the activity of this radionuclide. Six laboratories indicated that they looked for, but did not find the impurities in the solution. A comparison of the mean value of the measurements obtained by the laboratories that did not detect impurities showed no significant difference from that of the laboratories which did report impurity levels.

TABLE 1. LIST OF PARTICIPANTS, ALPHA-PARTICLE TEST SOLUTION
PLUTONIUM-239

Quality Assurance Program Eastern Environmental Radiation Facility P. O. Box 3009 Montgomery, AL 36109	Group H-8 University of California Los Alamos Scientific Laboratory P.O. Box 1663 Los Alamos, NM 87544
State of Connecticut Health Laboratories Environmental Chemistry Section 10 Clinton Street Hartford, CT 06101	Radiological Sciences Laboratory New York State Health Department 30 Russell Road Albany, NY 12206
Midwestern Facility Eberline Instrument Corporation 245 Roosevelt Road West Chicago, IL 60185	Mound Laboratory Monsanto Research Corporation P.O. Box 32 Miamisburg, OH 45342
State Hygienic Laboratory The University of Iowa Medical Laboratory Building Iowa City, IA 52242	State of Oregon Health Division Radiation Control Section 1400 S.W. 5th Avenue Portland, OR 97201
Winchester Engineering and Analytical Center U.S. Food & Drug Administration 109 Holton Street Winchester, MA 01890	Radiation Management Corporation 3508 Market Street Philadelphia, PA 19104
Nuclear Facilities and Contract Control Section Division of Radiological Health Bureau of Environmental and Occupational Health Michigan Department of Public Health 3500 N. Logan St., P.O. Box 30035 Lansing, MI 48909	South Carolina State Department of Health and Environmental Control 2600 Bull Street Columbia, SC 29201
Eberline Instrument Corporation Albuquerque Laboratory P.O. Box 3874 Albuquerque, NM 87110	Lynchburg Research Center P.O. Box 1260 Lynchburg, VA 24505
Group, H-5, Industrial Hygiene University of California Los Alamos Scientific Laboratory P.O. Box 1663, MS 486 Los Alamos, NM 87544	Radiochemistry Division United States Testing Company, Inc. 2800 George Washington Way Richland, WA 99352
	Laboratory of Radiation Ecology 104 Fisheries Center University of Washington Seattle, WA 98105

The order in which participants are listed in this table does not correspond to the order in which results are listed in Table 2 and Figure 1.

Six laboratories used alpha spectrometers and six laboratories used gas flow proportional counters. One laboratory used a zinc sulfide phosphor. Four laboratories did not indicate the type of detector they used. The mean value of the measurements obtained by those laboratories using alpha spectrometers was about the same as that value of those laboratories using gas flow proportional counters.

Four laboratories used plutonium-242 as a tracer. One laboratory used plutonium-236 as a tracer. Six laboratories indicated they electroplated their counting sources while five laboratories indicated they prepared their sources by evaporating the solution onto planchets. One laboratory precipitated the plutonium using lanthanum trifluoride (La F_3). The mean value of the measurements made by laboratories using tracers was not significantly different from that of laboratories that did not use tracers. There was no significant difference between the results of the laboratories that electroplated their sources and those that evaporated their sources.

RESULTS

The results reported by the 17 participants are given in Table 2 and Figure 1 as ratios of the participants reported mean values of alpha activity, corrected for decay to the date of calibration, to the EPA-calibrated activity value. This ratio is denoted as (X/EPA). For each X/EPA ratio presented in Figure 1, the linear sums of the random counting error at the 99 percent confidence level, plus the linear sums of the estimated limits of systematic error as reported by the participants, are shown as an error bar.

TABLE 2. RESULTS OF THE PLUTONIUM-239 MEASUREMENTS

Laboratory Code Letter	X/EPA Ratio	Uncertainty (%)	Laboratory Code Letter	X/EPA Ratio	Uncertainty (%)
A	1.020	5	J	1.028	3
B	1.007	7	K	1.002	10
C	0.894	*	L	1.025	6
D	0.823	6	M	1.153	7
E	1.037	2	N	1.103	4
F	0.932	17	O	4.563	6
G	1.049	10	P	1.439	*
H	1.111	6	Q	0.914	*
I	1.811	7			

* Uncertainties could not be determined for laboratories C, P and Q.

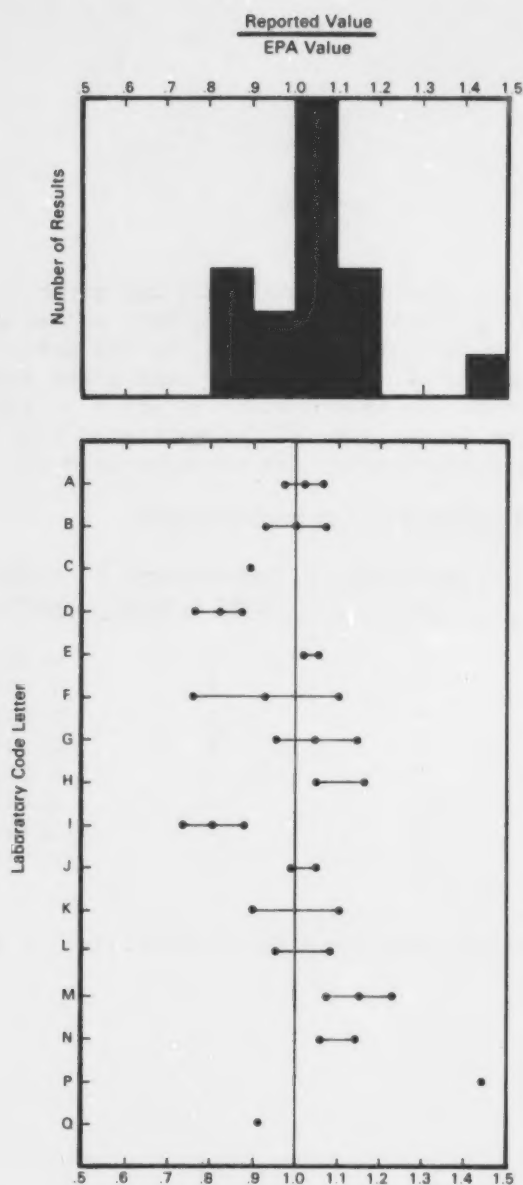


Figure 1. Reported results for the plutonium-239 radioactivity unknown solution. Participants in this intercomparison are listed in Table 1. The bars are total estimated errors (random plus estimated systematic errors). Uncertainties could not be determined for laboratories C, P and Q. Results from laboratory O are not included.

DISCUSSION

Twenty-eight laboratories received an alpha-particle test solution prepared by the National Bureau of Standards for use as a calibration standard. Fifteen laboratories reported values within $\pm 20\%$ of the EPA-certified value. Two laboratories data differed from the EPA-certified value by more than 20%, consequently, these data were not used in analyzing the results of this study. Comparisons between the various methodologies were made by examining the mean values and the standard deviation of each group. Inspection of these averages and standard deviations showed nothing significant. It was felt by the author that the sample size was so small that further statistical testing was not warranted.

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APPENDIX

U.S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS
WASHINGTON, D.C. 20234

REPORT OF CALIBRATION

PLUTONIUM-239

Alpha-Particle Solution Standard

prepared for

Environmental Protection Agency
Environmental Monitoring and Support Laboratory
Las Vegas, Nevada

This standard consists of carrier-free plutonium-239 in 4.1 grams of 5.0 molar nitric acid in a flame-sealed borosilicate-glass ampoule.

The total number of alpha particles emitted by plutonium-239 plus impurities per second per gram of solution on October 1, 1976 was

$*99.42 \pm 1.0\%*$

A master solution of carrier-free plutonium-239 was calibrated by depositing weighed aliquots on platinum disks and counting the dried deposits in the NBS "medium-geometry", defined-solid-angle ($\Omega \approx 0.4$ steradians), alpha-particle counter with scintillator detector. This standard was then prepared by quantitative mass dilutions of that master solution. Confirmatory measurements of the diluted solutions were made using both the "medium-geometry" counter and a liquid-scintillation counter. The measurements of the diluted and master solutions agreed to within 0.2%.

The uncertainty in the alpha-particle-emission rate, 1.0 percent, is the linear sum of 0.2 percent, which is the limit of the random error in the defined-solid-angle, alpha-particle-counter measurements of the master solution at the 99-percent confidence level ($3.17 S_m$, where S_m is the standard error computed from 11 determinations), and 0.8 percent, which is the linear sum of the estimated upper limits of conceivable systematic errors associated with these measurements and the dilutions.

Other isotopes of plutonium, as well as americium-241, are present in this solution standard. No attempt was made at NBS to determine the quantities of these impurities. However, the composition of the starting material has been extensively studied and intercompared and the reported composition is given on the reverse side of this report.

For the Director,

W. B. Mann
W. B. Mann, Chief
Radioactivity Section
Center for Radiation Research

December 8, 1976

Appendix (continued)

The composition of plutonium mixture Pu-1 circulated for intercomparison and used as the starting material for this standard is given below:

Isotope	Weight fraction on December 10, 1974 ^a	Laboratory ^b	Half life ^c (years)	Average relative α -particle emission rate on October 1, 1976 ^d
Pu-238	8.32×10^{-6}	LASL	87.79	0.0023
	8.45×10^{-6}	LLL		
Pu-239	0.992602	LASL	24146	1.0000
	0.992642	LLL		
Pu-240	7.311×10^{-3}	LASL	6537	0.0270
	7.263×10^{-3}	LLL		
Pu-241	9.028×10^{-5}	LASL	14.35	(0.1358 β^-)
	8.633×10^{-5}	LLL		
Am-241	6.1×10^{-7}	LASL	434.1	0.0004
	6.7×10^{-7}	LLL		
Total	1.000012	LASL		1.0297
	1.000000	LLL		

Note that, because the amount of americium-241 is still increasing and, to some extent, compensates for the decay of the plutonium, the total alpha-particle-emission rate will remain within 0.2% of the certified value for more than 100 years.

a. Date of purification.

b. LASL = Las Alamos Scientific Laboratory, Las Alamos, New Mexico.

LLL = Lawrence Livermore Laboratory, Livermore, California.

c. The half lives used are taken from appendix B, *American National Standard Calibration Techniques for the Calorimetric Assay of Plutonium - Bearing Solids Applied to Nuclear Materials Control*, N15.22 - 1975, except for the half life of plutonium-239, which is the value calculated from the composition data above and the measured total alpha-particle-emission rate.

d. Date of calibration.